

A neutron scattering study of the under-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, $x=0.09$ and 0.17 self-flux grown single crystals and the universality of the tricritical point

C. R. Rotundu,^{1,*} W. Tian,² K. C. Rule,³ T. R. Forrest,⁴ J. Zhao,⁴ J. L. Zarestky,² and R. J. Birgeneau^{4,5}

¹*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

²*Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, IA 50011, USA*

³*Helmholtz-Zentrum Berlin, Berlin, Germany*

⁴*Department of Physics, University of California, Berkeley, CA 94720, USA*

⁵*Department of Materials Science and Engineering, University of California, Berkeley, CA 94720, USA*

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We present a combination of elastic neutron scattering measurements in zero and 14.5 T and magnetization measurements in zero and 14 T on under-doped superconducting $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ $x=0.17$, and the same measurements in zero field on a non-superconducting crystal with $x=0.09$. The data suggest that the under-doped materials may not be electronic phase separated but rather have slightly inhomogeneous potassium doping. The temperature dependence of the magnetic order parameter (OP) below the transition of the sample with $x=0.09$ is more gradual than that for the case of the un-doped BaFe_2As_2 , suggesting that this doping may be in the vicinity of a tricritical point. We advance therefore the hypothesis that the tricritical point is a common feature of all superconducting 122s. For the $x=0.17$ sample, while T_c is suppressed from ≈ 17 K to ≈ 8 K by a magnetic field of 14 T, the intensity of the magnetic Bragg peaks (1 0 3) at 1.2 K is enhanced by 10% showing competition of superconductivity (SC) and antiferromagnetism (AFM). The intensity of the magnetic Bragg peaks (1 0 3) in the (T_c , T_N) temperature interval remain practically unchanged in 14.5 T within a 10% statistical error. The present results are discussed in the context of the existing literature.

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I. INTRODUCTION

High temperature superconductivity (HTSc) in the iron pnictides, with a T_c as high as 55 K for the case of $\text{SmFeAsO}_{1-\delta}$ and $\text{SmFeAsF}_x\text{O}_{1-x}$ ^{1,2}, is one of the most perplexing discoveries of the decade in the field of condensed matter physics. The 122 series (AFe_2As_2 , A = Ba, Sr, Ca, Eu) is of great interest since it is an oxygen-free HTSc. Superconductivity in the 122s can be induced by doping in any of the three atomic sites³⁻¹². The hole-doping achievable through chemical substitution with either K³, Na⁴ or Cs⁵ in the atomic site A can give a T_c as high as 39 K in the case of $\text{Ba}_{0.55}\text{K}_{0.45}\text{Fe}_2\text{As}_2$ ³. The antiferromagnetic (spin-density wave) and structural (tetragonal to orthorhombic) transitions that are near-coincident in the parent compounds^{13,14} are concomitantly and gradually suppressed upon doping. Although in the electron-doped BaFe_2As_2 the two transitions separate with doping¹⁵, it seems that there are examples pointing otherwise, as in the case of the isovalent ruthenium doped BaFe_2As_2 ^{16,17} and the case of electron-doped $\text{SrFe}_{2(1-x)}\text{Co}_{2x}\text{As}_2$ ¹⁸. The last is surprising if we consider the result on Sn-flux grown $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ crystals¹⁹ for which the two transitions are clearly separated. In the case of potassium (hole) doped BaFe_2As_2 ²⁰ the question of concomitant or separated transitions remains controversial. While powder neutron diffraction data argue for concomitant magnetic and structural transitions across the whole series^{20,21}, heat capacity on Sn-flux grown $\text{Ba}_{0.84}\text{K}_{0.16}\text{Fe}_2\text{As}_2$ single crystals shows two

distinctive peaks attributed by the authors to the magnetic and structural phase transitions, respectively²². In most cases the source of contradictory results appears to be connected to issues of sample quality. It has been pointed out that the proper flux to grow the 122s is FeAs ²³⁻²⁵, as other fluxes contaminate the sample with flux element inclusions, with a consequent impact on the physical properties. Neutron diffraction on powder BaFe_2As_2 ¹⁴ determined a first-order structural and magnetic transition. Complementary high resolution X-ray diffraction and heat capacity measurements on high quality BaFe_2As_2 crystals revealed a 1st order magnetic transition preceded by a structural transition that starts as a 2nd order transition at a slightly higher temperature but with a first order jump in the orthorhombic distortion coincident with the first order magnetic transition^{26,27}. For the electron-doped $\text{BaFe}_{2(1-x)}\text{Co}_{2x}\text{As}_2$ it has been shown recently that the magnetic transition order changes upon doping from 1st to 2nd through a tricritical point^{27,28}, which is believed to be relevant to the superconductivity phenomenon itself²⁹. For this series the structural transition is 2nd order. This seems to be different for the case of polycrystalline hole-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ of C. Avci *et al.*²⁰ for which both the magnetic and structural transitions are 1st order over the entire doping range. An early report on Sn-flux grown K-doped BaFe_2As_2 revealed an electronic phase separated material³⁰. A more recent atom probe tomography study on self-flux grown underdoped $\text{Ba}_{0.72}\text{K}_{0.28}\text{Fe}_2\text{As}_2$ provides evidence for a mixed scenario of phase coexistence and phase sep-

aration originating from variation of the dopant atom distributions³¹. In the present article we report complementary zero and 14.5 T elastic neutron scattering and zero and 14 T magnetization measurements on underdoped non-SC $x=0.09$ and SC $x=0.17$ ($T_c \approx 17$ K). For the non-SC $x=0.09$ sample the AFM transition is sharp (width within 1 K), consistent with a weakly 1st order transition, and the temperature dependence of the magnetic order parameter (OP) squared is more gradual than that for the case of the parent BaFe_2As_2 . This possibly indicates proximity to a tricritical point. For the higher doping sample $x=0.17$, the transition presents a distribution of T_N s due to a slight variation of the potassium dopant, leading to a rounding of the transition of about 6 K. This rounding makes it difficult to differentiate between first and second order behavior of the transition. Our neutron data show that, although the SC under-doped $x=0.17$ sample has a SC volume fraction of $\approx 40\%$, the downturn in the AFM order parameter below T_c and its enhancement in magnetic field provide possible evidence for microscopic coexistence of AFM and SC, similar to the case of the electron-doped 122s³².

II. EXPERIMENTAL PROCEDURE

Single crystals of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with potassium dopings x of 0.09, 0.17, 0.41, and 0.45³³ were synthesized by a self-flux method with details given in an earlier report³⁴. Samples with $x=0.41$ and 0.45 show no sign of AFM ordering, and are fully superconducting with $T_c=38$ K³⁴ and 39 K, $\Delta T_c=2$ K, respectively. The potassium doping has been extrapolated by comparing the c lattice determined from room temperature neutron data with c vs. x (K doping) data on crystals of H. Lou *et al.*³⁵ (Fig. 1). This data agree with c vs. x of the polycrystalline samples³⁶. Crystals of $\text{SrFe}_{2-x}\text{Ni}_x\text{As}_2$, $x=0.155$ were grown by self-flux method as well³⁷ and the precise Ni doping value was determined by inductively coupled plasma analysis.

Magnetization measurements on the K-doped samples were carried out using a Magnetic Property Measurement System (MPMS) and a Physical Property Measurement System (PPMS) from Quantum Design[®]. Resistivity measurements on the Ni-doped SrFe_2As_2 were performed in the PPMS. Zero field neutron diffraction measurements were performed at Oak Ridge National Laboratory (ORNL) with the High Flux Isotope Reactor's HB-1A triple axis spectrometer, using a horizontal collimation $40' - 40' - \text{sample} - 40' - 68'$ and fixed energy $E_i=14.6$ meV. The samples studied had K concentrations of $x=0.09$, 0.17, and 0.41 with masses of 19.5 mg, 45 mg, and 71.5 mg, respectively. In order to assess further sample quality, rocking curves of the (002) Bragg peak were recorded. For the $x=0.09$ sample the rocking curve showed two peaks that were separated by approximately 0.7° . Fitting these peaks to a Lorentzian squared profile gave FWHMs of 0.60° and 0.70° . For the

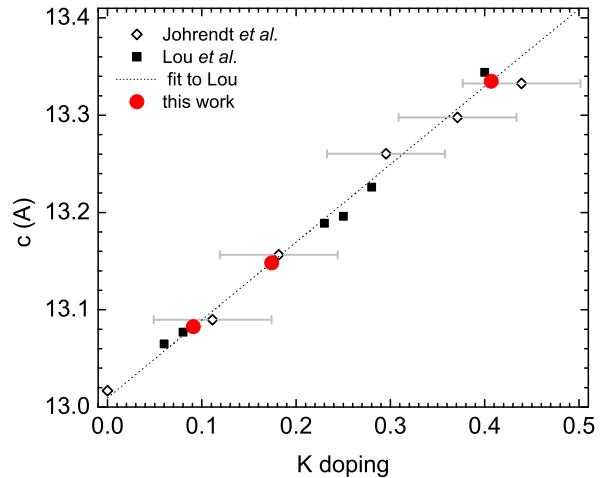


FIG. 1: c lattice versus x potassium doping, literature and present study data.

$x=0.17$ sample, the rocking curve of the same peak gave one main peak with a FWHM of 0.77° .

Neutron diffraction measurements of the $x=0.17$ sample of 50 mg in zero field and 14.5 T were performed at the Helmholtz-Zentrum Berlin (HZB) with a configuration of $60' - 20' - \text{sample} - 20'$ and fixed $E_f=5.0$ meV. The rocking curve of the (002) Bragg peak showed two peaks that were separated by 0.86° , both peaks had a FWHM of 0.47° .

For both ORNL and HZB neutron scattering experiments, the samples were mounted in a closed-cycle refrigerator and studied in the vicinity of the magnetic Bragg position $Q_{AFM}=(1\ 0\ 3)$. For all magnetization and neutron scattering measurements, the magnetic field was parallel with the (a b) crystallographic plane.

III. RESULTS AND DISCUSSION

The thermal evolution of the integrated intensity of the (1 0 3) magnetic Bragg peak in the non-SC $x=0.09$ sample is shown in Fig. 2, upper panel. In addition, the scattering from the same magnetic Bragg peak of the undoped parent compound BaFe_2As_2 is also shown (data is taken from Ref. 38). The peak intensity scales like the magnetic OP squared. For the sample $x=0.09$ the Néel temperature is 136 K. The figure shows that the magnetic OP squared in the $x=0.09$ sample evolves in a much more gradual manner than Wilson *et al.*'s data on $x=0$ ³⁸. Even so, there is a clear and sharp jump (within 1 K) of magnetic OP squared directly below the Néel temperature. This clearly shows that magnetic phase transition in this sample is still first order, albeit one that is weaker than the parent compound's. In critical phenom-

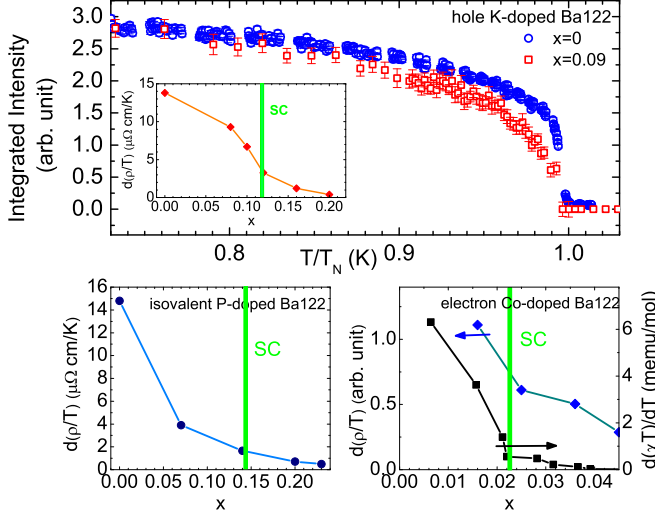


FIG. 2: The upper panel shows the integrated intensity of the (1 0 3) magnetic Bragg peak versus reduced temperature of the non-SC K-doped sample $x=0.09$ (\square) plotted against the same data for the un-doped $x=0$ (\circ) (from Ref. 38). The inset shows the magnitude of the $d\rho/dT$ peaks vs. x potassium doping of the series extracted from Ref. 39. Similar $d\rho/dT$ vs. x data is plotted for the isovalent phosphorus-doped BaFe_2As_2 in the lower left panel, and for the electron cobalt-doped in the lower right panel. The thick vertical lines indicate the doping corresponding to emergence of superconductivity in the series.

ena language this corresponds to a slight increase in the effective critical exponent describing the temperature dependence of the OP below the first order transition. We speculate that $x=0.09$ may be close to a tricritical point, similar to the one found in Co-doped BaFe_2As_2 ^{27,28}. It has been shown for the electron Co-doped BaFe_2As_2 ²⁸ that, around the tricritical point, the heat capacity C and $d(\chi T)/dT$ vs. doping present a change from a more abrupt variation (characteristic of a 1st order transition), to a monotonic and much slower variation (characteristic of a 2nd order transition). For the electron Co-doped BaFe_2As_2 in the lower right panel of Fig. 2, the magnitude of the peaks $d(\chi T)/dT$ vs. doping reproduced from Ref. 28 is drawn in comparison with the magnitude of $d\rho/dT$ ⁴⁰ peaks as extracted from Ref. 40. In the inset of the upper panel of Fig. 2 we plot the magnitude of the $d\rho/dT$ peaks vs. x of the same series, extracted from Ref. 39. The existence of an inflection point in the $d\rho/dT$ vs. x data may indicate a tricritical point at around $x \approx 0.12$ for the hole K-doped system. As found for the case of the Co(electron)-doped BaFe_2As_2 ²⁸, this tricritical point in K(hole)-doped BaFe_2As_2 is in the near proximity of emergence of superconductivity $0.125 \leq x \leq 0.133$ ²⁰. Finally, the lower left panel shows magnitude of $d\rho/dT$ peaks vs. phosphorus doping as extracted from Ref. 41⁴¹.

Figure 3 shows the integrated intensity of the (1 0 3)

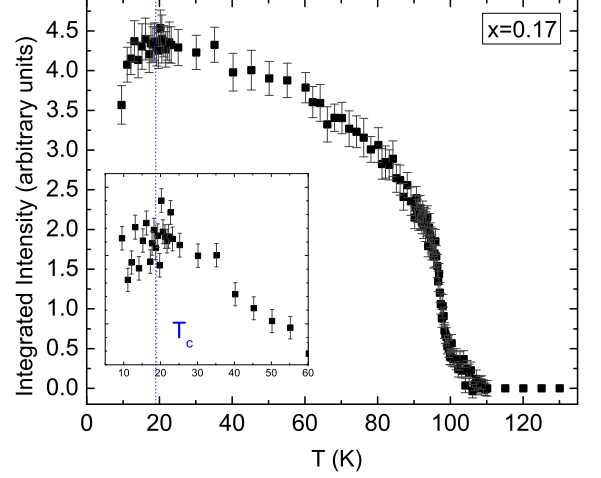


FIG. 3: Integrated intensity of the magnetic OP versus temperature for the sample $x=0.17$. The downturn of the OP below T_c shows coexistence of AFM and SC. In the inset it is the sum of counts versus temperature near T_c . The vertical interrupted line is a guide to the eye for T_c . The rounded transition at T_N is due to the slight K-doping inhomogeneity.

magnetic Bragg peak versus temperature of the sample $x=0.17$. The downturn of the intensity below T_c provides evidence for the microscopic coexistence of AFM and SC. In the inset is shown the sum of counts versus temperature near T_c . The downturn of the magnetic OP is less pronounced than for the case of homologous superconducting electron doped $\text{BaFe}_{2(1-x)}\text{Co}_x\text{As}_2$ ³² because of the low superconducting volume fraction. The “rounded” Néel transition (over a ≈ 6 K temperature range) is due in part to a slight distribution in the potassium doping, and therefore this will give an averaged $\langle T_N \rangle$. If we assume that the effect is due solely to a spread in doping, then the ≈ 6 K wide transition corresponds on the phase diagram³⁶ to a variation on potassium doping x of about 2.5%. The presence in the sample of small fractions of material with slightly smaller values of potassium doping will result in a non-zero magnetic order parameter above $\langle T_N \rangle$ and SC critical temperatures below $\langle T_c \rangle$ (untraceable by means of resistivity and magnetization measurements).

In order to investigate the effects of magnetic field on superconductivity, we have measured the magnetization of an $x=0.17$ sample. The onset of the diamagnetism as measured in 10 Oe is at ≈ 17 K (Fig. 4, right axis). The superconducting volume fraction is about 40%. This value is considerably higher than the 23 % reported for a higher potassium doped Sn-flux grown sample³⁰, and contrasts with the 98% value found by R. R. Urbano *et al.*²² in their Sn-flux grown $x=0.16$. In a magnetic field of 14 T (Fig. 4, left axis) T_c decreases to ≈ 8 K. This is expected since under-doped superconducting samples

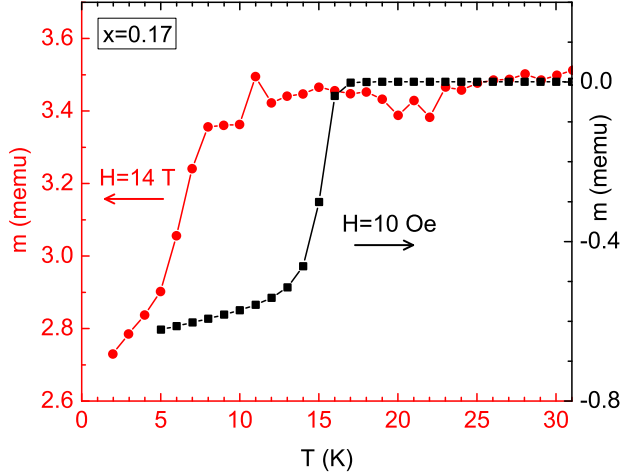


FIG. 4: Magnetization of the $x=0.17$ sample in $H=10$ Oe showing the onset of the diamagnetism at ≈ 17 K (right axis). In 14 T (left axis) T_c decreases to ≈ 8 K.

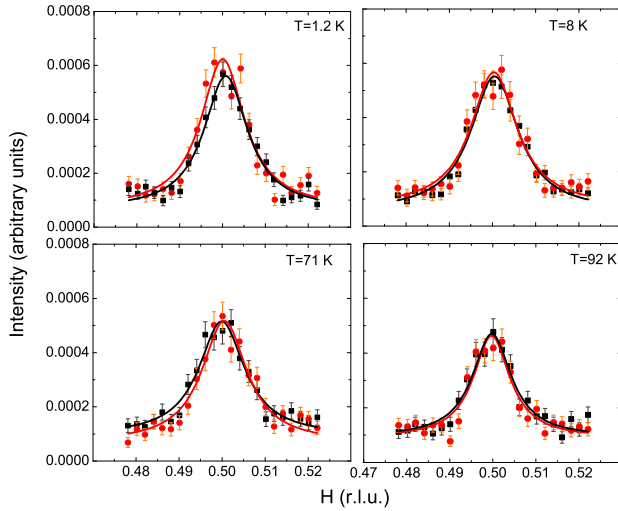


FIG. 5: Elastic neutron-scattering scans of the $(1\ 0\ 3)$ magnetic Bragg peak at different temperatures: $T=1.2, 8, 71$, and 92 K of the sample $x=0.17$. For each temperature the zero field data are indicated with filled squares and data in 14.5 T with filled circles, and the curves are Lorentzian fittings.

have a lower critical field H_{c2} than those that are optimally doped, where the critical field was estimated to be above 75 T⁴².

Figure 5 shows elastic neutron-scattering scans of the $(1\ 0\ 3)$ magnetic Bragg peak at different temperatures: $T=1.2, 8, 71$, and 92 K of the $x=0.17$ sample, in zero and 14.5 T. While for the 1.2 K the 14.5 T magnetic intensity is $\approx 10\%$ higher than for the zero field, our data

also show that a field of 14.5 T leaves the magnetic scattering practically unchanged within the errors for the (T_c, T_N) temperature range. This result certainly contrasts with the clear decrease of the magnetic intensity by $\approx 10\%$ in 13.5 T reported on Sn-flux grown higher potassium doped BaFe_2As_2 (with $T_c=32\pm 1$ K) of J. T. Park *et al.*³⁰. Therefore our 14.5 T data on the hole K-doped BaFe_2As_2 is similar to the 10 T high resolution neutron data on the electron underdoped $\text{BaFe}_{1.92}\text{Ni}_{0.08}\text{As}_2$ ($T_c=17$ K)⁴³. Here, below T_c the intensity of the magnetic $(1\ 1\ 3)$ peak is enhanced with $\approx 10\%$, while above T_c the intensity remains almost unchanged. One experiment to test the interplay between AFM and SC would be to determine whether or not a high magnetic field induces AFM in an optimally doped sample (without any trace of static AFM in zero field). This is very difficult to apply to the case of the optimally K-doped BaFe_2As_2 , as the critical field is over 75 T. Since for the case of the electron-doped 122s the critical field is much lower, we performed zero and in-field (14 T) resistivity measurements of optimally doped Ni-doped SrFe_2As_2 .

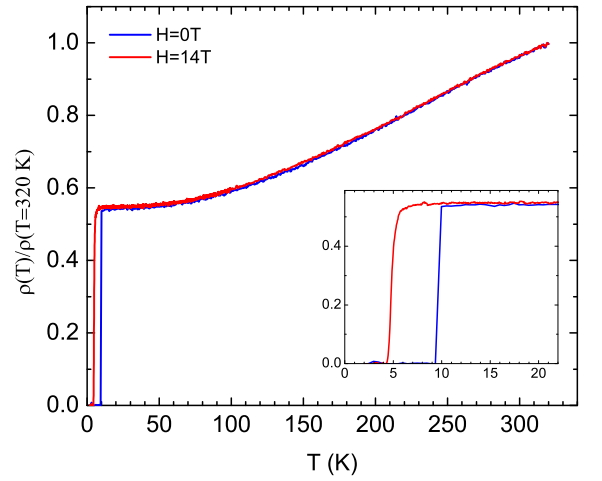


FIG. 6: Resistivity vs. temperature of the $\text{SrFe}_{2-x}\text{Ni}_x\text{As}_2$, $x=0.155$ normalized at its value at 320 K, in zero field and in 14 T// c . Before the measurement, the sample was annealed in low argon pressure for 24 hours at 700°C .

Figure 6 shows resistivity vs. temperature of the $\text{SrFe}_{2-x}\text{Ni}_x\text{As}_2$, with $x=0.155$ normalized at its value at 320 K value, in zero and in 14 T// c (it is known that for $H//c$ the critical field is lower than for $H//(a\ b)$ configuration⁴⁴). Before measurement, the sample was annealed in low argon gas pressure for 24 hours at 700°C ^{26,45}. Although T_c was suppressed from 10 to 5 K, there is no signature of any induced AFM in the 14 T// c data. It is important to mention that Ni-doped samples with T_c of 5 K are well into the coexistence of Sc and AFM region on the phase diagram, therefore exhibiting robust AFM. Our high field resistivity data are in

agreement therefore with neutron measurements in a 13.5 T//*c* field on optimally electron-doped BaFe_{1.9}Ni_{0.1}As₂ that showed that the field did not induce static AFM order⁴⁶. Therefore, part of the results of the hole- and electron-doped 122s seems to be consistent with a competing static AFM order and SC, similar to that for cuprate HTSc. The high field results reported here on the hole-doped Ba_{0.83}K_{0.17}Fe₂As₂ are similar to the case of cuprates for which the AFM order is strengthened with application of a magnetic field^{47,48}. Despite the resemblance of the shape of the phase diagrams^{50,51} for both iron pnictide and cuprate HTSc, the superconductivity in these materials appears to be of a different nature. We believe that these results will stimulate further exploration.

IV. SUMMARY

In summary, in the present article we report complementary elastic neutron scattering in zero and 14.5 T and magnetization measurements in zero and 14 T on under-doped SC *x*=0.17, and zero field on non-SC *x*=0.09. While for the non-SC *x*=0.09 sample the AFM transition is sharp consistent with a weakly 1st order transition, for higher doping *x*=0.17 the transition presents a broad distribution on *T_N* due to a slight variation of the K dopant. For sample *x*=0.09 the temperature dependence of the magnetic OP is more gradual than for the case of parent BaFe₂As₂, indicative of proximity to a tricritical point. This tricritical point seems to be an universal feature among all superconducting 122s. The

slight variation on the K dopant in the *x*=0.17 SC sample contributes to the fractional SC volume. Although the SC under-doped *x*=0.17 sample has a SC volume fraction of ≈40%, we were able to observe a downturn in the AFM order parameter below *T_c*, a clear sign of competition between AFM and SC, and similar to the one observed in the electron-doped 122s. As for the case of electron-doped 122s, a 14.5 T magnetic field enhances the AFM below *T_c* with ≈10%. This points, for the case at least of the 122s, toward a *s*[±] SC pairing symmetry³² in the hole-doped material, similar to that in the electron-doped 122s. Finally we mention that while writing the current paper, we became aware of related work by E. Wiesenmayer *et al.*⁵². Their combined X-ray and muon spin rotation on powder samples of the potassium under-doped materials show microscopical coexistence of AFM and SC.

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* Corresponding author: CRRotundu@lbl.gov

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